REMOVAL OF FLUORIDE FROM WATER SOLUTIONS BY NATURAL ZEOLITE MODIFIED WITH CATIONIC SURFACTANT

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ABSTRACT

The removal of fluoride from an aqueous solution by natural zeolite modified with a cationic surfactant was studied in a batch system. Hexadecyltrimethylammonium (HDTMA) bromide was used for zeolite modification. In the current study, the influence of several variables including pH of solution, initial concentration of fluoride, dose of surfactant-modified zeolite (SMZ), contact time, effects of competing ions, and effect of temperature were investigated. SMZ was characterized using XRD, XRF and SEM analyses. The equilibrium isotherm for fluoride uptake corresponded to the Langmuir model, and maximum fluoride adsorption capacity was found to be 0.96 mg/g. Kinetic studies have shown that the adsorption followed the pseudo-second-order model. When the pH was increased from 3 to 9, the fluoride removal efficiency decreased significantly. By increasing the concentration, the adsorption capacity of fluoride was increased. The best dosage of SMZ was found to be 0.2 g. The anion competition effect on fluoride adsorption by SMZ was very significant, as defluoridation in solutions containing Cl^-, I^-, and HCO^-_3 ions was higher than in those containing $H_2PO^-_4$, NO^-_3 and SO^{2-}_4 ions, and they were in the order of Cl^-> I^-> HCO^-_3> $H_2PO^-_4$> NO^-_3> SO^{2-}_4.

KEYWORDS:
Adsorption, fluoride removal, surfactant modified zeolite (SMZ)

1. INTRODUCTION

Excess fluoride in water causes health hazards to the natural environment. The high fluoride levels in drinking water and their impact on human health have increased the importance of fluoridation studies in many parts of the world. In groundwater, natural fluoride concentrations range from trace quantities to more than 2.5 mg/L [1]. In groundwater, the natural concentration of fluoride depends on the geological, chemical, and physical characteristics of the aquifer, the porosity and acidity of the soil and rocks, temperature, the action of other chemical elements, and the depth of wells. The maximum acceptable concentration of fluoride in water is 1.5 mg/L [1]. The major sources of fluoride in groundwater are fluoride-bearing rocks, such as fluorspar, cryolite, fluorapatite and hydroxylapatite. The fluoride content in the groundwater is a function of many factors, such as availability and solubility of fluoride minerals, velocity of flowing water, temperature, pH, and concentration of calcium and bicarbonate ions in water [2], and it enters the soil through weathering of rocks, precipitation or waste run off. Surface waters generally do not contain more than 0.3 mg/L of fluoride, unless they are polluted from external sources [3].

In Iran, fluoride is the major inorganic pollutant originating from groundwater. It is necessary to remove excessive fluoride from drinking water if its concentration is higher than 1.5 mg/L. To control fluoride concentrations in drinking water, several treatment options have been studied. Several defluoridation processes, such as adsorption, ion exchange, precipitation, electrodialysis, reverse osmosis, and electrochemical methods have been investigated to remove the excessive fluoride [4, 5]. Adsorption technology is frequently used as a robust technique to remove water-soluble ions from aqueous solutions, especially when these ions exist in low concentrations. Thus, a lot of studies have been reported on the use of various adsorbents for fluoride removal from drinking water. These studies have mainly been motivated by the need to have alternative low-cost adsorbents which have local availability; require little processing, and are superior in performance [6].

Natural zeolites are hydrated aluminosilicate materials having cage-like structure with internal and external surface areas of up to 1000 m^2/g and a cation exchange capacity of up to several milliequivalent per gram (meq/g). Most natural zeolites consist of a framework built up of SiO_4 and AlO_4 tetrahedra. Aluminum is in trivalent state and silicon is in tetravalent state, which makes them to be in negatively charged framework structures. Therefore, each mole of aluminum can produce one equivalent of cation exchange capacity to do a zeolite framework. As a matter of this characteristic, anions cannot be exchanged by zeo-

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